# First-Principles Molecular Dynamics (FPMD)

CASC Center for Applied Scientific Computing

## Overview

This project aims to develop high-performance software for First-Principles Molecular Dynamics (FPMD) simulations. First-principles (or *ab initio*) molecular dynamics is a microscopic description of matter that relies entirely on fundamental physical quantities and on the laws of quantum mechanics. The resulting approach is free of empirical assumptions specific to the system under study and is therefore applicable to a wide variety of problems, ranging from high-pressure physics to solvation chemistry, nanotechnology, and molecular biology.

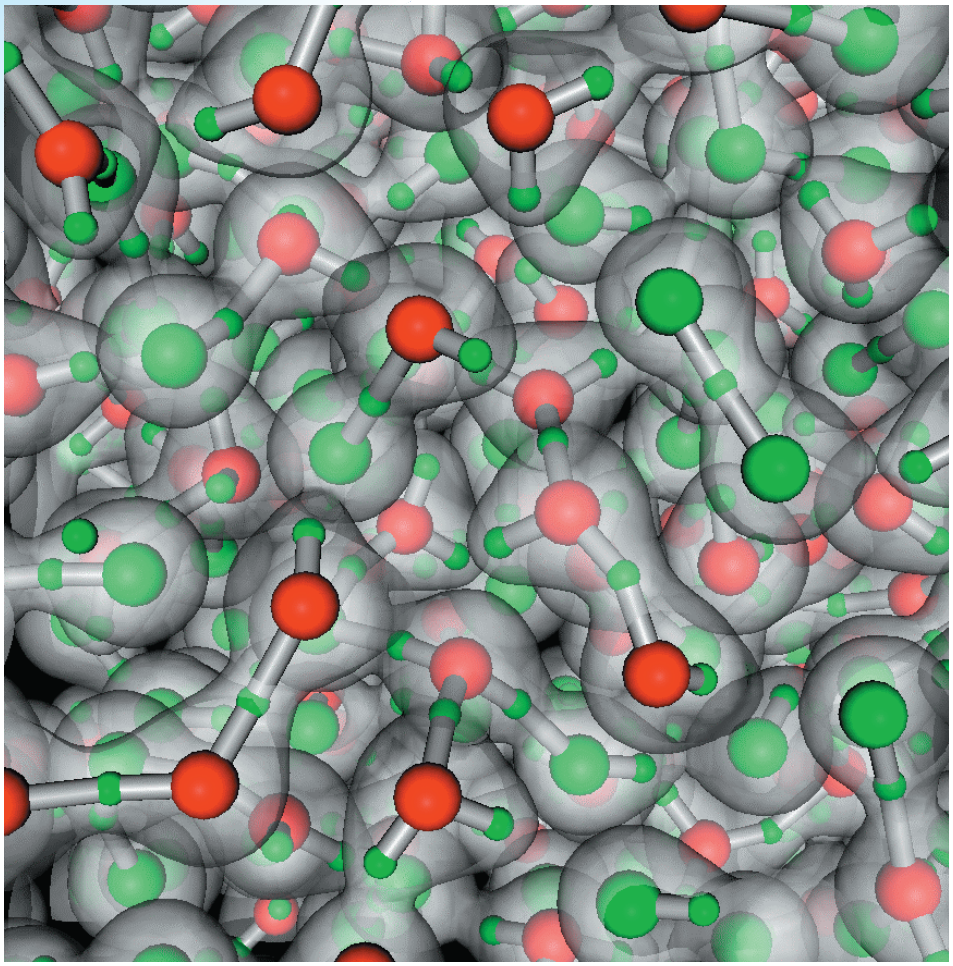

***Figure 1. Molecular structure of a mixture of hydrogen fluoride and water at high pressure and high temperature computed using the JEEP First-Principles Molecular Dynamics code. Large red and green spheres represent oxygen and fluorine atoms and small green spheres represent hydrogen atoms. The electronic density is shown as a semi-transparent isosurface.***

Molecular dynamics has been used for decades to investigate dynamical properties of molecules, solids, and liquids by numerical simulations. In the conventional (or *classical*) molecular dynamics approach, a model of interatomic interactions must be provided as input before a simulation can be carried out. Such models, or *interatomic potentials*, are based on *a priori* knowledge of the physical system studied. Ionic forces can be derived from such model potentials, and atomic trajectories are computed by integrating the Newtonian equations of motion. By contrast, First-Principles Molecular Dynamics does not require any *a priori* knowledge of interatomic interactions. Ionic forces are instead computed directly from the electronic structure of the system, which is obtained by solving an approximate form of the Schrödinger equation. This feature is important for simulations involving the formation and breaking of chemical bonds. Figure 1 shows an example of such a simulation where water and hydrogen fluoride were mixed under high pressure, leading to the formation of molecular complexes after the modification of several chemical bonds. In another application, the dynamics of DNA is investigated by simulating a fragment of the DNA backbone surrounded by water molecules (Figure 2). This type of simulation provides new insight into the structural properties of DNA and the mechanisms of its solvation in water. First-principles simulations also provide accurate information about the properties of materials subjected to extreme conditions (e.g., high temperature or high pressure) which are difficult to realize experimentally. This has allowed experimental physicists to interpret and complement experimental results, and sometimes guide the choice of new experiments.

## New Algorithms

First-Principles Molecule Dynamics simulations are computationally expensive and often require supercomputers just to simulate even a few hundred atoms. In most conventional implementations of FPMD, the cost of a simulation involving $N$ atoms is proportional to $N^3$, severely limiting

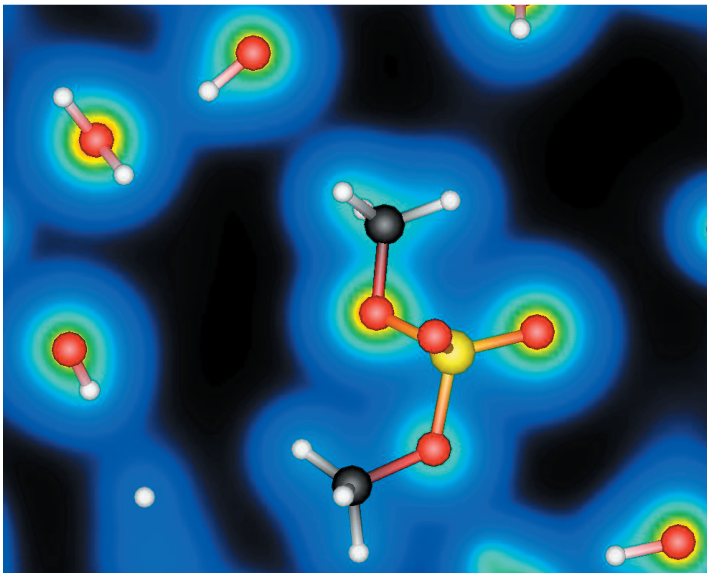

*Figure 2. A Dimethyl Phospate (DMP) molecule surrounded by water molecules. This simulation gives insight into the influence of water solvation on the structural properties of DNA.*

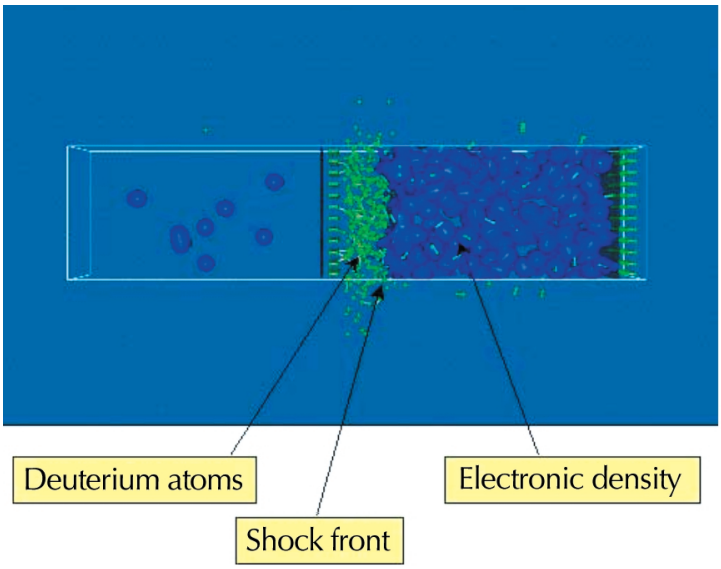


*Figure 3. Shock propagation in liquid deuterium. Deuterium atoms (green dots) are pushed at supersonic velocity by an array of deuterium molecules. The formation of the shock front is observed on the atomic scale. The electronic density is represented by the blue isosurface.*

large-scale simulations. An important area of research pursued within this project consists of exploring new algorithms that increase the efficiency of FPMD simulations. This is done both by efficiently exploiting the massively parallel architecture of large computers and by devising approximations that reduce the computational cost from $O(N^3)$ to $O(N)$, i.e., linear scaling. Such an algorithm based on a domain-decomposition, real-space, finite-difference formulation was recently implemented on LLNL's computers and will considerably extend the range of feasible simulations.

## Scalable Software

JEEP is a scalable parallel code developed within this project that implements First-Principles Molecular Dynamics within the Density Functional, plane-wave pseudopotential formalism. JEEP has been used to simulate the properties of systems as diverse as molecular liquids, solids, and semiconductor surfaces. In a recent large-scale simulation, JEEP was used to simulate the propagation of a shock in liquid deuterium (Figure 3). It provided detailed insight into the electronic and structural properties of the shock front. This simulation involved 1320 deuterium atoms and ran on 2640 processors of the ASCI White computer installed at LLNL.

## Collaborations

We are collaborating with the Physics and Advanced Technologies (PAT) directorate as well as the Biology and Biotechnology Research Program (BBRP) at LLNL.

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